Spin and orbital ferromagnetism in strongly correlated itinerant-electron systems

V. Yu. Irkhin* and M. I. Katsnelson
Institute of Metal Physics, 620219 Ekaterinburg, Russia and
Institute for Molecules and Materials, Radboud University Nijmegen, 6525 ED Nijmegen, The Netherlands

Spectra of one-particle and collective excitations in narrow-band ferromagnets with unquenched orbital moments are calculated in various theoretical models. The interaction of spin and orbital excitations with conduction electrons results in the damping of the former which, however, turns out to be rather small; therefore, apart from usual spin waves, well-defined orbitons can exist. Non-quasiparticle states occur in the electron energy spectrum near the Fermi energy due to this interaction. The criteria of stability of the saturated spin and orbital ferromagnetic ordering are considered. Possible effects of orbital ordering in magnetite and in colossal magnetoresistance manganites are discussed.

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I. INTRODUCTION

Orbital ordering in strongly correlated systems is a subject of interest for a rather long period [1], this interest growing considerably last time (see, e.g., recent papers [2, 3, 4, 5, 6]). For orbitally ordered magnets the energy spectrum is more rich than for purely spin-moment magnetic materials since it contains additional excitation branches ("orbitons"). Relevance of the orbital degrees of freedom for physics of the colossal magnetoresistance (CMR) manganites [3, 4, 7, 8, 9] is one of the reasons for the revival of this field. A large orbital contribution to the total magnetic moment has been observed recently by the X-ray magnetic circular dichroism (XMCD) method for magnetite Fe_3O_4 [10]. The latter substance (the most widespread natural iron compound) is probably one of the most intriguing magnetic oxides because of controversial experimental information concerning its ground state and so called "Verwey transition" [12, 13] (see also a discussion in Ref.14). The magnetite is also a promising material: it is a half-metallic ferromagnet (more exactly, ferrimagnet) with a high Curie temperature and perspectives of applications in spintronics [15]. Since current theoretical description of the half-metallic ferromagnetism (for a review, see Refs.15, 16) ignores the orbital degrees of freedom, it should be generalized to include them and to take into account probable orbital ferromagnetism in the magnetite and, possibly, in other strongly correlated conducting ferromagnets. Such a generalization is the aim of the present work.

The paper is organized as follows. In Section 2 we formulate the models of a narrow-band ferromagnet including both spin and orbital degrees of freedom and discuss their relevance for the colossal magnetoresistance (CMR) manganites and for the magnetite. In Section 3 the Bose-type Green's functions are calculated which determine the spectrum and damping of spin waves (magnons) and orbitons. In Section 4 the Fermi-type one-particle Green's functions are derived and peculiarities of the electron energy spectrum owing to the electron-magnon and electron-orbiton interactions are considered. Using the expressions obtained the conditions for stability of saturated spin and orbital ferromagnetic ordering are investigated. In Section 5 a general picture of the half-metallic ferromagnetism in the presence of the orbital degrees of freedom is discussed.

II. MODELS OF A CONDUCTING FERROMAGNET WITH ORBITAL DEGREES OF FREEDOM

Previous works concerning the role of the orbital degrees of freedom in strongly correlated magnetic systems treated mainly the insulator case, in particular, orbital ferromagnetism accompanied with spin antiferromagnetism [1], orbital antiferromagnetism [5], or orbital liquid [2]. In Ref.11 a mean-field phase diagram of an insulator including orbital and spin ferromagnetism region was obtained. In contrast with these works, we consider here *conducting* ferromagnets with both spin and orbital degrees of freedom.

We start with the many-electron system with two ground terms of the d^n and d^{n+1} configurations, $\Gamma_n = \{SL\}$ and $\Gamma_{n+1} = \{S'L'\}$. The corresponding periodic Anderson model that describes the electron hopping with the transitions between two these states only reads

$$\mathcal{H} = \mathcal{H}_0 + \sum_{\mathbf{k}\sigma m} t_{\mathbf{k}m} c^\dagger_{\mathbf{k}\sigma m} c_{\mathbf{k}\sigma m} + \sum_{\mathbf{k}im\sigma} \left(V_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma m} e^{i\mathbf{k}\mathbf{R}_i} a_{il\sigma m} + \text{h.c.} \right) \tag{1}$$

where $\mathcal{H}_0$ is the Hamiltonian for strongly correlated d-electrons described by the operators $a_{il\sigma m}$, $t_{\mathbf{k}m}$ is the band energy for conduction electrons, $V_{\mathbf{k}}$ is the matrix element of hybridization. It is suitable to use the representation of

the Fermi operators for the localized electrons in terms of the many-body atomic quantum numbers [17]

$$a^{\dagger}_{il\sigma m} = (n+1)^{1/2} \sum G^{S'L'}_{SL} C^{L'\mu'}_{L\mu,lm} C^{S'M'}_{SM,\frac{1}{2}\sigma} X_i \left(S'L'M'\mu', SLM\mu\right) \tag{2}$$

where $G^{S'L'}_{SL}$ are the fractional parentage coefficients, $X_i(\alpha\beta) = |i\alpha\rangle\langle i\beta|$ are the many-electron Hubbard operators [18] which satisfy the relations at a site i

$$X_i(\alpha\beta)X_i(\gamma\varepsilon) = \delta_{\beta\gamma}X_i(\alpha\varepsilon), \ \sum_{\alpha} X_i(\alpha\alpha) = 1. \tag{3}$$

By a canonical Schrieffer-Wolff transformation the model (1) is reduced to the broad-band $s-d$ exchange model with the exchange parameter $I_{sd} \sim V^2/\Delta$ (Δ is the difference of the term energies). In a general case, we have the $s-d$ exchange model with two magnetic configurations. This model was used to investigate the electron spectrum and calculate the Kondo temperature in Refs.17, 19. Here we introduce a further simplification assuming that only one of the competing configurations has non-zero orbital moment $L = l$. This assumption holds for the magnetite which has d^5 and d^6 ground-state configurations for Fe^{3+} and Fe^{2+} respectively, the first configuration having zero orbital moment. A similar situation takes place for the CMR manganites (with d^3 and d^4 configuration for Mn^{4+} and Mn^{3+}): due to the relevance of $t_{2g} - e_g$ crystal-field splitting the former configuration corresponds to the completely filled t_{2g} band with $L = 0$.

According to Refs.17, 19 the $s-d$ exchange Hamiltonian takes the form

$$\mathcal{H}_{sd} = -I_{sd} \sum C^{S'M'}_{SM,\frac{1}{2}\sigma} C^{S'M'}_{SM'',\frac{1}{2}\sigma'} X_i(SLMm, SLM''m) c^{\dagger}_{i\sigma m} c_{i\sigma' m}. \tag{4}$$

Rearranging the product of the Clebsch-Gordan coefficients C with the use of $6j$-symbols,

$$\sum_{M'} C^{S'M'}_{SM,\frac{1}{2}\sigma} C^{S'M'}_{SM'',\frac{1}{2}\sigma'} = \frac{3\sqrt{2}}{2} \left(\frac{2S'+1}{2S+1}\right)^{1/2} \left\{ \begin{array}{ccc} 1/2 & 1/2 & 1 \\ S & S' & S' \end{array} \right\} \sum_{q=-1}^{1} C^{SM}_{SM'',1q} C^{\frac{1}{2}\sigma}_{\frac{1}{2}\sigma',1-q}, \tag{5}$$

and taking into account that $C^{SM}_{SM'',1q}$ are the matrix elements of spin operators, we obtain the $s-d$ exchange Hamiltonian with orbital degrees of freedom in the form

$$\mathcal{H} = \sum_{\mathbf{k}\sigma m} t_{\mathbf{k}m} c^{\dagger}_{\mathbf{k}\sigma m} c_{\mathbf{k}\sigma m} - I \sum_{i\sigma\sigma' m} \mathbf{S}_i \boldsymbol{\sigma}_{\sigma\sigma'} c^{\dagger}_{i\sigma m} c_{i\sigma' m} + \mathcal{H}_d \tag{6}$$

where $\mathcal{H}_d$ is the Heisenberg Hamiltonian of the localized-spin system, $\boldsymbol{\sigma}$ are the Pauli matrices, the $s-d$ exchange parameter I is renormalized in comparison with I_{sd} in Eq.(4). A similar one-impurity model was discussed for the Mn^{2+} ion (orbital singlet problem) [20, 21].

In contrast with the case of rare-earth elements where the orbital moments are not quenched, but act as a part of the total orbital moment (the Russel-Saunders coupling [17, 19, 22]), for $3d$-electron systems we have to consider the case of strong crystal field where the excitation spectrum is characterized by spin and orbital quantum numbers separately. However, in the case under consideration the quantum number m enters the model in a very simple way (the Hamiltonian is diagonal in the orbital indices) and can label both spherical and cubic harmonics. Of course, a relatively simple form of the Hamiltonian (6) results from our assumption that only one configuration has a non-zero orbital moment. Fortunately, it is the case that is most interesting for real systems, as it was mentioned above. Thus a complicated mathematics involving the orbital Clebsch-Gordan coefficients is relevant only when we have *two* many-electron terms with $L \neq 0$.

Note that in the case of $j-j$ coupling (e.g., $5f$-electron systems) where the electron states $|jm\rangle$ are characterized by the projection of the momentum j, we have the Hamiltonian

$$\begin{aligned} \mathcal{H} &= \sum_{\mathbf{k}m} t_{\mathbf{k}m} c^{\dagger}_{\mathbf{k}m} c_{\mathbf{k}m} - I \sum_{i} \mathbf{J}_i \mathbf{j}_i, \\ j^{\alpha}_i &= \sum_{mm'} C^{jm'}_{jm,1\alpha} c^{\dagger}_{im} c_{im'}. \end{aligned} \tag{7}$$

Provided that the crystal field Hamiltonian is diagonal in m, the model (6) does not mix (at least in the lowest orders of perturbation theory) the states with different m, see Section 4. Therefore we focus in the present work mainly on a more rich and complicated narrow-band case (which is also most interesting for real transition metal compounds). This case should be described by a two-configuration Hubbard model where both conduction electrons

and local moments belong to the same d-band, the states with $n+1$ electrons playing the role of current-carrier states. After performing the procedure of mapping onto the corresponding state space, the one-electron Fermi operators for the strongly correlated states $a^{\dagger}_{il\sigma m}$ are replaced by many-electron operators according to Eq.(2). Taking into account the values of the Clebsh-Gordan coefficients which correspond to the coupling of momenta S and $1/2$ we obtain

$$\mathcal{H} = \sum_{\mathbf{k}\sigma m} t_{\mathbf{k}m} g^{\dagger}_{\mathbf{k}\sigma m} g_{\mathbf{k}\sigma m}. \tag{8}$$

Here we have redefined the band energy by including the many-electron renormalization factor,

$$t_{\mathbf{k}m}(n+1)(G^{S'0}_{SL})^2/(2l+1) \rightarrow t_{\mathbf{k}m}, \tag{9}$$

and

$$\begin{aligned} g^{\dagger}_{i\sigma m} &= \sum_{M=-S}^{S} \sqrt{\frac{S-\sigma M}{2S+1}} X_i(S-1/2, M+\frac{\sigma}{2}; SMm), \quad S' = S-1/2, \\ g^{\dagger}_{i\sigma m} &= \sum_{M=-S}^{S} \sqrt{\frac{S+\sigma M+1}{2S+1}} X_i(S+1/2, M+\frac{\sigma}{2}; SMm), \quad S' = S+1/2 \end{aligned} \tag{10}$$

where $|SMm\rangle$ are the empty states with the orbital index m, $|S'M'\rangle$ are the singly-occupied states with the total on-site spin $S' = S \pm 1/2$ and its projection M', $\sigma = \pm$.

We see that the two-configuration Hamiltonian is a generalization of the narrow-band $s-d$ exchange model with $|I| \rightarrow \infty$ [17, 23]. In the case where the configuration d^{n+1} has larger spin than the configuration d^n, we have the effective "$s-d$ exchange model" with $I > 0$, and in the opposite case with $I < 0$; it is worthwhile to remind that our band energy is renormalized according to (9) in comparison with the $s-d$ exchange Hamiltonian (cf. the factor of $1/2$ for the narrow-band Hubbard model without orbital degeneracy [23]).

In the case of $j-j$ coupling we have

$$g^{\dagger}_{i\sigma m} \rightarrow g^{\dagger}_{im} = \sum_{MM'} C^{J'M'}_{JM,jm} X_i(J'M', JM)$$

The specification of the Hamiltonian (8) for the case of a saturated ferromagnetic state (where only maximum spin projections give a contribution) reads for $I < 0$ ($S' = S - 1/2$)

$$\mathcal{H} = \sum_{\mathbf{k}m} t_{\mathbf{k}m} \left[\frac{2S}{2S+1} X^{a,+m}_{-\mathbf{k}} X^{+m,a}_{\mathbf{k}} + \frac{1}{2S+1} X^{a,-m}_{-\mathbf{k}} X^{-m,a}_{\mathbf{k}} + \frac{\sqrt{2S(2S-1)}}{2S+1} \left(X^{a,+m}_{-\mathbf{k}} X^{-m,b}_{\mathbf{k}} + \text{h.c.} \right) \right]. \tag{11}$$

Here $|+m\rangle$ and $|-m\rangle$ are the on-site states without excess electrons and with localized spin projection S and $S-1$, respectively; $|a\rangle = |S-1/2, S-1/2\rangle$ and $|b\rangle = |S-1/2, S-3/2\rangle$ are the states with one excess electron and the total spin projection $S-1/2$ and $S-3/2$, respectively; $X^{\alpha\beta}_{\mathbf{k}}$ are the Fourier transforms of the Hubbard operators $X_i(\alpha, \beta)$.

For the case where $I > 0$ ($S' = S + 1/2$) we have

$$\mathcal{H} = \sum_{\mathbf{k}m} t_{\mathbf{k}m} \left[X^{u,+m}_{-\mathbf{k}} X^{+m,u}_{\mathbf{k}} + \frac{2S}{2S+1} X^{v,-m}_{-\mathbf{k}} X^{-m,v}_{\mathbf{k}} + \frac{1}{2S+1} X^{v,+m}_{-\mathbf{k}} X^{+m,v}_{\mathbf{k}} + \sqrt{\frac{2S}{2S+1}} \left(X^{u,+m}_{-\mathbf{k}} X^{-m,v}_{\mathbf{k}} + \text{h.c.} \right) \right] \tag{12}$$

where $|u\rangle = |S+1/2, S+1/2\rangle$ and $|v\rangle = |S+1/2, S-1/2\rangle$ are the singly-occupied states.

We will consider also the simplest model where one of the configuration corresponding to the "current carriers" has zero spin and orbital moments (non-magnetic holes):

$$\mathcal{H} = \sum_{\mathbf{k}\sigma m} t_{\mathbf{k}m} X^{0,\sigma m}_{-\mathbf{k}} X^{\sigma m,0}_{\mathbf{k}}. \tag{13}$$

This is a formal generalization of the standard narrow-band Hubbard model in the X-operator representation to include the orbital degeneracy. Apart from the m-dependence of the band energy $t_{\mathbf{k}m}$, the Hamiltonian (13) includes spin and orbital degrees of freedom on equal footings. However, for finite Hubbard repulsion U the symmetry of spin and charge channels is in fact lost owing to the Hund interaction, so that the state with orbital ferromagnetism

and spin antiferromagnetism can become favorable in comparison with the spin antiferromagnetic state [1]. Note the equivalence of the model (13) with the replacement $t_{\mathbf{k}m} \to t_{\mathbf{k}m}/2$ and the model (11) for $S = 1/2$.

For orbitally degenerate case the subbands $t_{\mathbf{k}m}$ are connected by the point-group symmetry transformations and have identical densities of states. However, in real situations usually the Jahn-Teller lattice instability takes place which destroys the exact orbital degeneracy [1]. Due to the Jahn-Teller distortions, both additional on-site crystal-field splitting arises (which means the shift of the centers of the subbands) and hopping integrals are changed (which means different $\mathbf{k}$-dependences). Thus we will assume further that all the functions $t_{\mathbf{k}m}$ are in general different.

III. COLLECTIVE SPIN AND ORBITAL EXCITATIONS

The spectrum of electron and spin excitations in the above-discussed narrow band models can be calculated similar to the papers [23, 25, 26], the difference being in the occurrence of transitions into orbital states.

Even in the narrow band case where small interaction parameter is absent we have an hierarchy of energy scales owing to both formal expansion parameters (inverse nearest-neighbor number $1/z$, quasiclassical parameter $1/S$) and small current-carrier concentration. Therefore the width of the "magnon" band is much smaller than characteristic electron energies. A similar approach was developed earlier for degenerate ferromagnetic semiconductors [23].

First we calculate the retarded commutator Green's function describing spin and orbital excitations in the simplest model (13),

$$G^{\sigma\sigma'}_{\mathbf{q}mm'}(\omega) = \langle\langle X^{\sigma m,\sigma' m'}_{\mathbf{q}} | X^{\sigma' m',\sigma m}_{-\mathbf{q}} \rangle\rangle_\omega, \, \mathrm{Im}\omega > 0.$$

To derive the equation of motion, we use the commutation rules for X -operators, which follow from Eq.(3). We obtain

$$\omega G^{\sigma\sigma'}_{\mathbf{q}mm'}(\omega) = N_{\sigma m} - N_{\sigma' m'} + \sum_{\mathbf{k}} (t_{\mathbf{k}-\mathbf{q}m'} - t_{\mathbf{k}m}) \langle\langle X^{0,\sigma' m'}_{\mathbf{q}-\mathbf{k}} X^{\sigma m,0}_{\mathbf{k}} | X^{\sigma' m',\sigma m}_{-\mathbf{q}} \rangle\rangle_\omega. \tag{14}$$

In the next equation of motion we make a simple decoupling described in Ref.25, which corresponds to the first-order term in $1/z$-perturbation theory (each order in $1/z$ corresponds formally to a summation over an additional wave vector):

$$\begin{aligned}(\omega - E_{\mathbf{k}\sigma m} + E_{\mathbf{k}-\mathbf{q}\sigma' m'}) \langle\langle X^{0,\sigma' m'}_{\mathbf{q}-\mathbf{k}} X^{\sigma m,0}_{\mathbf{k}} | X^{\sigma' m',\sigma m}_{-\mathbf{q}} \rangle\rangle_\omega &= n_{\mathbf{k}\sigma m} - n_{\mathbf{k}-\mathbf{q}\sigma' m'} \\ &+ (t_{\mathbf{k}m} n_{\mathbf{k}\sigma m} - t_{\mathbf{k}-\mathbf{q}m'} n_{\mathbf{k}-\mathbf{q}\sigma' m'}) G^{\sigma\sigma'}_{\mathbf{q}mm'}(\omega). \end{aligned} \tag{15}$$

Here the "Hubbard-I" energies $E_{\mathbf{k}\sigma m}$ and the occupation numbers $n_{\mathbf{k}\sigma m}$ are given by the "Hubbard-I" [28] expressions

$$\begin{aligned} E_{\mathbf{k}\sigma m} &= t_{\mathbf{k}m}(N_{\sigma m} + N_0), \; N_\alpha = \langle X^{\alpha\alpha}_i \rangle, \\ n_{\mathbf{k}\sigma m} &= \langle X^{0,\sigma m}_{-\mathbf{k}} X^{\sigma m,0}_{\mathbf{k}} \rangle = (N_{\sigma m} + N_0) f(E_{\mathbf{k}\sigma m}), \end{aligned} \tag{16}$$

$f(E)$ being the Fermi function. Then we have

$$\begin{aligned} G^{\sigma\sigma'}_{\mathbf{q}mm'}(\omega) &= \left(N_{\sigma m} - N_{\sigma' m'} + \sum_{\mathbf{k}} \frac{(t_{\mathbf{k}-\mathbf{q}m'} - t_{\mathbf{k}m})(n_{\mathbf{k}\sigma m} - n_{\mathbf{k}-\mathbf{q}\sigma' m'})}{\omega - E_{\mathbf{k}\sigma m} + E_{\mathbf{k}-\mathbf{q}\sigma' m'}} \right) \\ &\times \left(\omega - \sum_{\mathbf{k}} \frac{(t_{\mathbf{k}-\mathbf{q}m'} - t_{\mathbf{k}m})(t_{\mathbf{k}m} n_{\mathbf{k}\sigma m} - t_{\mathbf{k}-\mathbf{q}m'} n_{\mathbf{k}-\mathbf{q}\sigma' m'})}{\omega - E_{\mathbf{k}\sigma m} + E_{\mathbf{k}-\mathbf{q}\sigma' m'}} \right)^{-1}. \end{aligned} \tag{17}$$

In the saturated ferromagnetic state ($N_{\sigma m} = \delta_{\sigma +} N_m$) and for small hole concentrations $c = N_0$ the pole of the Green's function (17) yields a simple result for the spin-wave frequency

$$\omega^{+-}_{\mathbf{q}mm'} = \sum_{\mathbf{k}} (t_{\mathbf{k}-\mathbf{q}m'} - t_{\mathbf{k}m}) f(t_{\mathbf{k}m}). \tag{18}$$

In the case of saturated orbital ordering ($N_{\sigma m} = \delta_{m+} N_\sigma$) the orbital excitation frequency $\omega^{\sigma\sigma'}_{\mathbf{q}+m'}$ ($m = + \neq m'$) is given by the same expression (18). Provided that the states m' and m are split by the crystal field, there is a gap in the corresponding orbiton spectrum branch. Assuming for simplicity that the band bottom corresponds to Γ-point $\mathbf{k} = 0$, we derive

$$\omega^{\sigma\sigma'}_{\mathbf{q}+m'} \simeq c(t_{\mathbf{q}m'} - t_{0m'}) + c\Delta, \; \Delta = t^{\min}_{m'} - t^{\min}_{+}. \tag{19}$$

It should be noted that the gap $\omega_0 = c\Delta$ is proportional to the current carrier concentration, but not just equal to the crystal-field splitting Δ, in contrast with the case of weak crystal field typical for the rare-earth systems [29]. Actually, in our case characteristic orbiton energies for finite wave vectors have the same order of magnitude as the magnon energies.

The magnon damping can be obtained similar to Ref.23 by calculating higher-order terms in $1/z$,

$$\gamma_{\mathbf{q}mm'}(\omega) = \pi \sum_{\mathbf{k}\mathbf{p}m''} (t_{\mathbf{k}-\mathbf{q}m'} - t_{\mathbf{k}m})^2 [n_{\mathbf{k}-\mathbf{q}+\mathbf{p}m''}(1-n_{\mathbf{k}m}) + N_B(\omega_{\mathbf{p}})(n_{\mathbf{k}-\mathbf{q}+\mathbf{p}m''} - n_{\mathbf{k}m})]\delta(\omega + t_{\mathbf{k}m} - t_{\mathbf{k}-\mathbf{q}+\mathbf{p}m''} - \omega_{\mathbf{p}})$$
$$= \pi \sum_{\mathbf{k}\mathbf{p}m''} (t_{\mathbf{k}-\mathbf{q}m'} - t_{\mathbf{k}m})^2 (\omega_{\mathbf{p}} - \omega) \frac{\partial n_{\mathbf{k}m}}{\partial t_{\mathbf{k}m}} [N_B(\omega_{\mathbf{p}}) - N_B(\omega_{\mathbf{p}} - \omega)]\delta(t_{\mathbf{k}m} - t_{\mathbf{k}-\mathbf{q}+\mathbf{p}m''}) \quad (20)$$

with $N_B(\omega_{\mathbf{p}})$ being the Bose function. The damping turns out to be finite at $T = 0$, unlike the case of the Heisenberg ferromagnet. However, it is proportional to the frequency ω and contains formal small parameters, which leads to very small value [23]. Thus both spin-flip and non-spin-flip orbital excitations are well defined. This fact is non-trivial in a narrow-band case (a similar situation takes place also in the antiferromagnetic case, cf. Ref.5).

In the model (8) the results for the "magnon" spectrum are more complicated and depend in a non-trivial way on the spin value. The calculations for the ferromagnetically saturated state can be performed similar to Ref.23. For the case $S' = S - 1/2$ we write down the sequence of equations of motion

$$\omega G^{+-}_{\mathbf{q}mm'}(\omega) = N_m + \frac{1}{2S+1} \sum_{\mathbf{k}} [(t_{\mathbf{k}-\mathbf{q}m'} - 2St_{\mathbf{k}m}) \langle\langle X^{a,-m'}_{\mathbf{q}-\mathbf{k}} X^{+m,a}_{\mathbf{k}} | X^{-m',+m}_{-\mathbf{q}} \rangle\rangle_\omega$$
$$+ \sqrt{2S(2S-1)} t_{\mathbf{k}m'} \langle\langle X^{a,+m'}_{-\mathbf{k}} X^{+m,b}_{\mathbf{k}+\mathbf{q}} | X^{-m',+m}_{-\mathbf{q}} \rangle\rangle_\omega]. \quad (21)$$

The equation for the first Green's function in the right-hand side is obtained as above (see Eq.(15)), and for the second one we have

$$(\omega - t^*_{\mathbf{k}m'} N_m) \langle\langle X^{a,+m'}_{-\mathbf{k}} X^{+m,b}_{\mathbf{k}+\mathbf{q}} | X^{-m',+m}_{-\mathbf{q}} \rangle\rangle_\omega = \sqrt{2S(2S-1)} \langle X^{a,+m}_{-\mathbf{k}} X^{+m,a}_{\mathbf{k}} \rangle t_{\mathbf{k}m'} G^{+-}_{\mathbf{q}mm'}(\omega) \quad (22)$$

where

$$t^*_{\mathbf{k}m} = \frac{2S}{2S+1} t_{\mathbf{k}m}. \quad (23)$$

Then the excitation energy in the leading order in $1/z$ reads

$$\omega^{+-}_{\mathbf{q}mm'} = \frac{1}{2S} \sum_{\mathbf{k}} [(t^*_{\mathbf{k}-\mathbf{q}m'} - 2St^*_{\mathbf{k}m}) f(t^*_{\mathbf{k}m}) + (2S-1) t^*_{\mathbf{k}m'} f(t^*_{\mathbf{k}m'})] \quad (24)$$

(more strict calculations in the absence of orbital degeneracy [23] yield a closed integral equation for the magnon spectrum). At the same time, the frequency of the non-spin-flip orbital transitions is obtained similar to Eq.(18) and is given for a saturated orbital-ordered state by the expression

$$\omega^{++}_{\mathbf{q}mm'} = \sum_{\mathbf{k}} (t^*_{\mathbf{k}-\mathbf{q}m'} - t^*_{\mathbf{k}m}) f(t^*_{\mathbf{k}m}), \quad (25)$$

so that a dependence on S is absent.

For $S' = S + 1/2$ we have the equations of motion

$$\omega G^{+-}_{\mathbf{q}mm'}(\omega) = N_m - \frac{1}{2S+1} \sum_{\mathbf{k}} t_{\mathbf{k}m} \langle\langle X^{u,-m'}_{\mathbf{q}-\mathbf{k}} X^{+m,u}_{\mathbf{k}} - \sqrt{\frac{2S}{2S+1}} X^{u,+m}_{-\mathbf{k}} X^{+m',v}_{\mathbf{k}+\mathbf{q}} | X^{-m',+m}_{-\mathbf{q}} \rangle\rangle_\omega, \quad (26)$$

$$(\omega - t_{\mathbf{k}m} N_m) \langle\langle X^{u,-m'}_{\mathbf{q}-\mathbf{k}} X^{+m,u}_{\mathbf{k}} | X^{-m',+m}_{-\mathbf{q}} \rangle\rangle_\omega = -t_{\mathbf{k}m} \langle X^{u,+m}_{-\mathbf{k}} X^{+m,u}_{\mathbf{k}} \rangle G^{+-}_{\mathbf{q}mm'}(\omega), \quad (27)$$

$$[\omega + t_{\mathbf{k}m} N_m - t_{\mathbf{k}+\mathbf{q}m'} N_{m'}/(2S+1)] \langle\langle X^{u,+m}_{-\mathbf{k}} X^{+m',v}_{\mathbf{k}+\mathbf{q}} | X^{-m',+m}_{-\mathbf{q}} \rangle\rangle_\omega = \sqrt{\frac{2S}{2S+1}} \langle X^{u,+m}_{-\mathbf{k}} X^{+m,u}_{\mathbf{k}} \rangle G^{+-}_{\mathbf{q}mm'}(\omega). \quad (28)$$

Then we derive

$$\omega^{+-}_{\mathbf{q}mm'} = \sum_{\mathbf{k}} \frac{t_{\mathbf{k}+\mathbf{q}m'}N_{m'} - t_{\mathbf{k}m}N_m}{2St_{\mathbf{k}m}N_m + t_{\mathbf{k}m}N_m - t_{\mathbf{k}+\mathbf{q}m'}N_{m'}} t_{\mathbf{k}m} f(t_{\mathbf{k}m}). \tag{29}$$

The orbital frequency for a saturated state reads

$$\omega^{++}_{\mathbf{q}mm'} = \sum_{\mathbf{k}} (t_{\mathbf{k}+\mathbf{q}m'} - t_{\mathbf{k}m}) f(t_{\mathbf{k}m}). \tag{30}$$

It should be mentioned that, instead of the difference $t_{\mathbf{k}m}N_m - t_{\mathbf{k}+\mathbf{q}m'}N_{m'}$, a resolvent occurs in the denominator in the right-hand side of Eq.(29) at more rigorous calculations which do not use the $1/z$-expansion (cf. Ref.23). In particular, in the case of ferromagnetically saturated spin and orbital ordering (all the sites without excess electrons are in the orbital state $|+m\rangle = |++\rangle$ with some $m \equiv +$) we obtain the following exact result for the magnon pole ($m = m'$):

$$\omega^{+-}_{\mathbf{q}m} = \sum_{\mathbf{k}} \frac{t_{\mathbf{k}+\mathbf{q}m} - t_{\mathbf{k}m}}{2S + (t_{\mathbf{k}m} - t_{\mathbf{k}+\mathbf{q}m}) R_{\mathbf{k}m}(\omega^{+-}_{\mathbf{q}m})} f(t_{\mathbf{k}m}), \tag{31}$$

$$R_{\mathbf{k}m}(\omega) = \sum_{\mathbf{p}} \frac{1 - f(t_{\mathbf{k}+\mathbf{p}m})}{\omega + t_{\mathbf{k}m} - t_{\mathbf{k}+\mathbf{p}m} - \omega^{+-}_{\mathbf{p}m}}. \tag{32}$$

Note that the imaginary part of the expression (31) describes the magnon damping.

IV. ELECTRON SPECTRUM AND INSTABILITIES OF THE SATURATED STATE

In the broad-band $s-d$ exchange model (6) we can perform a decoupling in spirit of a ladder approximation (cf. Ref.30) to obtain the Green's function for the case of zero temperature $T = 0$

$$\langle\langle c_{\mathbf{k}\sigma m} | c^{\dagger}_{\mathbf{k}\sigma m} \rangle\rangle_E = [E - t_{\mathbf{k}\sigma m} - \Sigma^{\sigma}_{\mathbf{k}m}(E)]^{-1} \tag{33}$$

where $t_{\mathbf{k}\sigma m} = t_{\mathbf{k}m} - \sigma IS$ is the mean-field (Hartree-Fock) spectrum. The electron self-energy reads

$$\Sigma^{\sigma}_{\mathbf{k}m}(E) = \frac{2I^2 S R^{\sigma}_{\mathbf{k}m}}{1 + \sigma I R^{\sigma}_{\mathbf{k}m}}, \tag{34}$$

$$R^{\uparrow}_{\mathbf{k}m}(E) = \sum_{\mathbf{q}} \frac{n^{\downarrow}_{\mathbf{k}-\mathbf{q}m}}{E - t_{\mathbf{k}-\mathbf{q}\downarrow m} + \omega_{\mathbf{q}}}, R^{\downarrow}_{\mathbf{k}}(E) = \sum_{\mathbf{q}} \frac{1 - n^{\uparrow}_{\mathbf{k}-\mathbf{q}m}}{E - t_{\mathbf{k}-\mathbf{q}\uparrow m} - \omega_{\mathbf{q}}} \tag{35}$$

with $n^{\sigma}_{\mathbf{k}m} = f(t_{\mathbf{k}\sigma m})$. Imaginary part of (34) describes the non-quasiparticle states which are due to non-pole contributions (branch cut of the Green's function) [15, 24]. In the saturated ferromagnetic case $n^{\downarrow}_{\mathbf{k}m} \neq 0$ for $I < 0$ and $n^{\uparrow}_{\mathbf{k}m} \neq 0$ for $I > 0$, and these states with $\sigma = -\mathrm{sign}I$ lie below (above) the Fermi level, respectively. The corresponding densities of states for a twofold orbital-degenerate band are shown in Figs.1,2. One can see that in the model (6) the subbands with different m are uncoupled (we do not consider their mixing owing to off-diagonal crystal field), so that the picture of the half-metallic ferromagnetism state discussed in Refs.15, 24 is not qualitatively changed, although the number of excitation branches increases.

A more complicated situation takes place for the narrow-band limit, a formal reason being in occurrence of inter-subband transitions owing to non-trivial commutation relations for many-electron X-operators. First we consider the one-particle spectrum in the simplest model (13). We treat the case $T = 0$ and restrict ourselves to the case of spin- and orbital-saturated ferromagnetic state (all the singly occupied on-site states have the spin projection $\sigma = +$ and the orbital state $m \equiv +$). Since this ground state is non-degenerate, the one-particle spectrum at small hole concentrations $c = N_0$ can be investigated in a strict way similar to Ref.23. We have to calculate the retarded anticommutator Green's function

$$G_{\mathbf{k}\sigma m}(E) = \langle\langle X^{\sigma m,0}_{\mathbf{k}} | X^{0,\sigma m}_{-\mathbf{k}} \rangle\rangle_E, \qquad \mathrm{Im}E > 0. \tag{36}$$

The current carriers with $|\sigma m\rangle = |++\rangle$ propagate as free ones,

$$G_{\mathbf{k}++}(E) = (E - t_{\mathbf{k}+})^{-1}. \tag{37}$$

In the case $|\sigma m\rangle \neq |{+}{+}\rangle$, to extract the operators of spin and orbital excitations, it is convenient to use the kinematical relation which follows from Eq.(3)

$$X^{\sigma m,0}_{\mathbf{k}} = \sum_{\mathbf{q}} X^{\sigma m,++}_{\mathbf{q}} X^{++,0}_{\mathbf{k}-\mathbf{q}}. \tag{38}$$

Then we have

$$G_{\mathbf{k}\sigma m}(E) = \sum_{\mathbf{q}} F_{\mathbf{k}\mathbf{q}\sigma m}(E), \quad F_{\mathbf{k}\mathbf{q}\sigma m}(E) = \langle\langle X^{\sigma m,++}_{\mathbf{q}} X^{++,0}_{\mathbf{k}-\mathbf{q}} | X^{0,\sigma m}_{-\mathbf{k}} \rangle\rangle_E. \tag{39}$$

After a natural decoupling procedure (cf. Ref.24) we obtain the integral equation for the function F

$$(E - t_{\mathbf{k}-\mathbf{q}+} + \omega^{+\sigma}_{\mathbf{q}+m}) F_{\mathbf{k}\mathbf{q}\sigma m}(E) = n_{\mathbf{k}-\mathbf{q}}[1 - (t_{\mathbf{k}-\mathbf{q}+} - t_{\mathbf{k}m}) \sum_{\mathbf{p}} F_{\mathbf{k}\mathbf{p}\sigma m}(E)] \tag{40}$$

where $n_{\mathbf{k}} = \langle X^{0,++}_{-\mathbf{k}} X^{++,0}_{\mathbf{k}} \rangle = f(t_{\mathbf{k}+})$. Solving this equation we derive

$$G_{\mathbf{k}\sigma m}(E) = \frac{1}{E - t_{\mathbf{k}m} + 2\overline{S}/R_{\mathbf{k}\sigma m}(E)} \tag{41}$$

where $2\overline{S} = 1 - c$ is the average spin magnetization,

$$R_{\mathbf{k}\sigma m}(E) = \sum_{\mathbf{q}} \frac{n_{\mathbf{k}-\mathbf{q}}}{E - t_{\mathbf{k}-\mathbf{q}+} + \omega^{+\sigma}_{\mathbf{q}+m}}. \tag{42}$$

Thus the Green's function with minority spin (orbital) projection has non-pole structure, the non-quasiparticle states below the Fermi level being of crucial importance to satisfy the sum rule

$$\begin{aligned} \int dE f(E) g_{\sigma m}(E) &= \sum_{\mathbf{k}} \langle X^{0,\sigma m}_{-\mathbf{k}} X^{\sigma m,0}_{\mathbf{k}} \rangle = N_0, \\ g_{\sigma m}(E) &= -\frac{1}{\pi} \mathrm{Im} \sum_{\mathbf{k}} G_{\mathbf{k}\sigma m}(E), \end{aligned} \tag{43}$$

irrespective of σ, m.

The spectrum picture is presented in Fig.3. To simplify numerical calculations, we average the resolvent $R_{\mathbf{k}\sigma m}(E)$ in (41) over $\mathbf{k}$,

$$R_{\mathbf{k}\sigma m}(E) \to \overline{R}_{\sigma m}(E) = \int d\omega K_{\sigma m}(\omega) \sum_{\mathbf{k}} \frac{n_{\mathbf{k}}}{E - t_{\mathbf{k}+} + \omega}$$

(this approximation yields the correct behavior near the Fermi level, cf. Ref.26, although yields an irrelevant unphysical shift of the band bottom by the maximum magnon frequency). We use the semielliptic magnon density of states $K_{\sigma m}(\omega)$ which is proportional (with the corresponding shift) to the bare electron density of states according to (19), the band edge ω_0 being determined by the crystal-filed splitting. When neglecting spin (orbital) dynamics, $\mathrm{Im}R_{\mathbf{k}\sigma m}(E)$ has a jump at the Fermi level. For gapless magnons $\mathrm{Im}\overline{R}_{\sigma m}(E)$ (this quantity is also shown in Fig.3) vanishes at E_F according to the law $(E_F - E)^{3/2}$ [24]. The "Kondo" singularity of $\mathrm{Re}\overline{R}_{\sigma m}(E)$ owing to the Fermi function $n_{\mathbf{k}}$ results in that the dependence of $g_{\sigma m}(E)$ near E_F is considerably more sharp than just of $\mathrm{Im}\overline{R}_{\sigma m}(E)$. The contributions of orbitons with a gap spectrum demonstrate a threshold energy equal to the gap, that is, in the presence of the orbital splitting $\mathrm{Im}\overline{R}_{\sigma m}(E)$ starts from $E_F - \omega_0$. At the same time, the threshold energy decreases strongly for the total density of states $g_{\sigma m}(E)$. Besides that, the orbital splitting results in a height increase of the $g_{\sigma m}(E)$ peak below the Fermi level.

The instability of the saturated state ($N_{\sigma m} \neq 0$ for $|\sigma m\rangle \neq |{+}{+}\rangle$) with increasing c corresponds to occurrence of a non-zero spectral density above E_F since

$$\int dE[1 - f(E)] g_{\sigma m}(E) = N_{\sigma m}. \tag{44}$$

This spectral density comes from the quasiparticle pole determined by the equation

$$R_{\mathbf{k}\sigma m}(E_F) = \frac{2\overline{S}}{\max t_{\mathbf{k}m} - E_F}. \tag{45}$$

The analytical estimations are complicated due to the logarithmic singularity in the resolvent $R_{\mathbf{k}\sigma m}(E)$. It should be noted that a simple cutoff of this singularity at a characteristic magnon frequency (cf. Ref. [27]) is insufficient since the energy dependence of the resolvent plays an important role ($\mathrm{Im}R_{\mathbf{k}\sigma m}(E) \propto (E_F - E)^{3/2}$ and the maximum in $\mathrm{Re}R_{\mathbf{k}\sigma m}(E)$ does not correspond to E_F). Thus an integration with the magnon spectral density is required. As it was demonstrated by the corresponding numerical calculations for the Hubbard model [26], the critical concentration of holes c_{crit} makes up about 20-30% for a number of crystal lattices. To calculate the second critical concentrations c'_{crit} (a transition from the non-saturated ferromagnetism into the paramagnetic state), more advanced approximations taking into account longitudinal spin fluctuations are necessary [26]. Note that in a more realistic case of finite on-site interaction the conditions for a ferromagnetically saturated state can be considerably more strict.

Provided that the spectra $t_{\mathbf{k}m}$ with different m are connected by crystal symmetry transformations (e.g., states corresponding to the irreducible t_{2g} representation in a cubic field), orbital and spin instabilities of the saturated state coincide. However, if the orbital subbands are split by the crystal field Δ (or, in other words, an orbital pseudomagnetic field is present), the criteria for the orbital and spin-flip instabilities can become different owing to the difference in the spectra $t_{\mathbf{k}m}$ and $t_{\mathbf{k}+}$ and the gap in the spectrum $\omega^{+\sigma}_{\mathbf{q}+m}$.

The analytical calculations of the one-particle excitation spectrum for the model (11) are performed in a similar way. We restrict ourselves again to the case of a small conduction electron concentration c. In the case $S' = S - 1/2$ spin-down electrons have the spectrum $t^*_{\mathbf{k}+}$ (see Eq.(23)). For the function

$$F_{\mathbf{kq}}(E) = \langle\langle X^{-,++}_{\mathbf{q}} X^{++,a}_{\mathbf{k}-\mathbf{q}} | X^{a,-+}_{-\mathbf{k}} \rangle\rangle_E \tag{46}$$

we obtain after some simplifications the integral equation

$$(E - t^*_{\mathbf{k}-\mathbf{q}+} + \omega^{+-}_{\mathbf{q}++}) F_{\mathbf{kq}}(E) = n^*_{\mathbf{k}-\mathbf{q}} [1 - \frac{1}{2S}(t^*_{\mathbf{k}-\mathbf{q}+} - t^*_{\mathbf{k}+}) \sum_{\mathbf{p}} F_{\mathbf{kp}}(E)] \tag{47}$$

with $n^*_{\mathbf{k}} = f(t^*_{\mathbf{k}+})$. Then we derive

$$G_{\mathbf{k}\uparrow+}(E) = \langle\langle X^{-+,a}_{\mathbf{k}} | X^{a,-+}_{-\mathbf{k}} \rangle\rangle_E = \frac{2S}{E - t^*_{\mathbf{k}+} + (2S - c)/R^*_{\mathbf{k}\sigma+}(E)} \tag{48}$$

where $R^*_{\mathbf{k}\sigma m}(E)$ differs from (42) by the replacement $t_{\mathbf{k}+} \to t^*_{\mathbf{k}+}$. The condition for the spin-flip instability corresponds to occurrence of the pole of the expression (48) above the Fermi level. As follows from numerical estimations, such a condition can be hardly satisfied for $S > 1/2$ (although the magnon frequency is also more soft in this case because of the factor $(2S)^{-1}$ in Eq.(24)).

The orbital instability is essentially governed by the Hamiltonian of the structure (13) (the first term in Eq.(11)) and is determined by the Green's function

$$\langle\langle X^{+m,a}_{\mathbf{k}} | X^{a,+m}_{-\mathbf{k}} \rangle\rangle_E = \frac{1}{E - t^*_{\mathbf{k}m} + (1 - c)/R^*_{\mathbf{k}+m}(E)}. \tag{49}$$

Since in such a case the factor of $2S$ at $1/R^*(E)$ is absent, orbital ordering becomes unstable at some electron concentrations, and we can treat the state with saturated ferromagnetic spin ordering, but destroyed orbital ordering.

In the case $S' = S + 1/2$ (model (12)) spin-up electrons behave as free ones, and we have for the spin-down Green's function

$$[E - t_{\mathbf{k}+}/(2S+1)] \langle\langle X^{++,v}_{\mathbf{k}} | X^{v,++}_{-\mathbf{k}} \rangle\rangle_E = 1 - c + \sqrt{\frac{2S}{2S+1}} \sum_{\mathbf{q}} t_{\mathbf{k}-\mathbf{q}+} L_{\mathbf{kq}}(E), \tag{50}$$

$$L_{\mathbf{kq}}(E) = \langle\langle X^{++,-+}_{\mathbf{q}} X^{++,u}_{\mathbf{k}-\mathbf{q}} | X^{v,++}_{-\mathbf{k}} \rangle\rangle_E, \tag{51}$$

$$\begin{aligned}(E - t_{\mathbf{k}-\mathbf{q}+} - \omega_{\mathbf{q}}) L_{\mathbf{kq}}(E) &= \sqrt{\frac{2S}{2S+1}} t_{\mathbf{k}-\mathbf{q}+} \langle\langle X^{++,v}_{\mathbf{k}} | X^{v,++}_{-\mathbf{k}} \rangle\rangle_E \\ &\quad - \sum_{\mathbf{p}} t_{\mathbf{k}-\mathbf{p}+} (1 - n_{\mathbf{k}-\mathbf{p}}) L_{\mathbf{kp}}(E)\end{aligned} \tag{52}$$

where $n_{\mathbf{k}} = \langle X^{u,++}_{-\mathbf{k}} X^{++,u}_{\mathbf{k}} \rangle = f(t_{\mathbf{k}+})$, $\omega_{\mathbf{q}} = \omega^{+-}_{\mathbf{q}++}$. Solving this system we obtain the expression which also has a typically "non-quasiparticle" form (without quasiparticle poles, at least, at not too large current carrier concentration)

$$\langle\langle X^{++,v}_{\mathbf{k}} | X^{v,++}_{-\mathbf{k}} \rangle\rangle_E = \frac{2S+1}{E - t_{\mathbf{k}+} + 2S/\widetilde{R}_{\mathbf{k}}(E)} \tag{53}$$

$$\widetilde{R}_{\mathbf{k}}(E) = \sum_{\mathbf{q}} \frac{1 - n_{\mathbf{k}-\mathbf{q}}}{E - t_{\mathbf{k}-\mathbf{q}+} - \omega_{\mathbf{q}}}. \tag{54}$$

Note that the result (53) is rigorous for the empty conduction band (spin-polaron problem) and is in agreement with (33)-(35) at $I \to +\infty$. As follows from the spectral representation, $n^{**}_{\mathbf{k}} \equiv \langle X^{v,++}_{-\mathbf{k}} X^{++,v}_{\mathbf{k}} \rangle = 0$ (provided that the Green's function (53) has no poles) since $\mathrm{Im}\widetilde{R}_{\mathbf{k}}(E < E_F) = 0$ because of the factor $1 - n_{\mathbf{k}-\mathbf{q}}$. Physically, there is no spin-flip instability in the $s-d$ model with large $I > 0$ since spin-down electrons are not present in the ground state. The corresponding spectrum picture is shown in Fig.4

Note that the Green's function (53) itself does not determine an instability of the saturated state since the spectral representation gives

$$-\frac{1}{\pi}\int dE[1 - f(E)]\mathrm{Im}\sum_{\mathbf{k}} \langle\langle X^{\sigma m,v}_{\mathbf{k}} | X^{v,\sigma m}_{-\mathbf{k}} \rangle\rangle_E = N_{\sigma m}. \tag{55}$$

To treat the instability we have to consider the Green's function with $\sigma = -$,

$$\langle\langle X^{-m,v}_{\mathbf{k}} | X^{v,-m}_{-\mathbf{k}} \rangle\rangle_E = \sum_{\mathbf{q}} \langle\langle X^{-m,++}_{\mathbf{q}} X^{++,v}_{\mathbf{k}-\mathbf{q}} | X^{v,-m}_{-\mathbf{k}} \rangle\rangle_E, \tag{56}$$

which is proportional to the distribution function $n^{**}_{\mathbf{k}}$ and is expressed in terms of the resolvent

$$R^{**}_{\mathbf{k}+m} = \sum_{\mathbf{q}} \frac{n^{**}_{\mathbf{k}-\mathbf{q}}}{E - t^{**}_{\mathbf{k}-\mathbf{q}} + \omega_{\mathbf{q}}} \tag{57}$$

where $t^{**}_{\mathbf{k}}$ is the spectrum of new quasiparticles which can occur with increasing electron concentration.

The orbital instability is determined by the first term in the Hamiltonian (12),

$$\langle\langle X^{+m,u}_{\mathbf{k}} | X^{u,+m}_{-\mathbf{k}} \rangle\rangle_E = \frac{1}{E - t_{\mathbf{k}m} + (1-c)/R_{\mathbf{k}+m}(E)}, \tag{58}$$

and is again S-independent.

V. CONCLUSIONS

In this paper we have presented the picture of excitation spectrum in a saturated conducting (half-metallic) ferromagnet with orbital degrees of freedom within the framework of simple many-electron models. It is proven that the half-metallic ferromagnetic state does exist in the presence of orbital degeneracy. We have focused our consideration on strongly correlated systems with pronounced term (multiplet) effects. In such a situation the description that uses atomic many-electron quantum numbers is most adequate; formally it is provided by the Green's function method for the Hubbard X-operators.

In contrast with usual itinerant-electron ferromagnets, additional collective excitation branches (orbitons) occur. Due to the smallness of the current carrier concentrations, these modes possess low frequencies in comparison with typical electron energies such as the crystal-field splitting. Typically, the orbiton energies are of the same order of magnitude as the magnon energies. Also, mixed excitations with the simultaneous change of spin and orbital projections exist ("optical magnons"). All these excitations can be well defined in the whole Brillouin zone, the damping due to the interaction with current carriers being small enough.

We have calculated one-particle Green's functions in the non-degenerate saturated ferromagnetic state. The expressions obtained yield different criteria for spin and orbital instabilities. It turns out that the saturated spin ferromagnetism is more stable than the orbital one in the realistic case $S > 1/2$ (e.g., for magnetite and for colossal magnetoresistance manganites). This means that the half-metallic ferromagnetic phases both with saturated and non-saturated orbital moments can arise. A more detailed investigation with the use of real band and spectrum is required for concrete cases where orbital degrees of freedom yield important contributions to magnetic properties. In some situations (e.g., near the quantum phase transitions like those in virtual ferroelectrics [31]) the orbital modes can become soft, which should result in orbital instabilities at smaller conduction electron concentrations c.

Due to the electron-orbiton interaction, additional contributions to the non-quasiparticle density of states can arise. Since the magnons are gapless, the electron-magnon contribution leads to a rather sharp one-sided increase of the density of states (with a crossover energy scale of the order of a typical magnon energy $\overline{\omega}$) starting just from the

Fermi energy, $\delta g(E) \propto (|E_F - E|/\overline{\omega})^{3/2}$ [15, 23, 24]. In the case of half-metallic ferromagnets with unquenched orbital moment one can predict additional contributions to $g(E)$ which correspond to orbital channels. Probably, the most simple way to probe the density of non-quasiparticle states is to measure the spin-polarized tunneling current [32, 33, 34]. Therefore it would be very interesting to investigate the density of states of "suspicious" materials by, e.g., spin-polarized scanning tunneling microscopy (STM) technique [35], the non-quasiparticle states being observable below the Fermi energy for the case of magnetite and above it for the case of colossal magnetoresistance manganites. It is worthwhile to mention that the non-quasiparticle states should exist also at the surface of the half-metallic ferromagnets [36] since STM is a surface-sensitive method.

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[*] E-mail: Valentin.Irkhin@imp.uran.ru

[1] K. I. Kugel and D. I. Khomskii, Usp. Fiz. Nauk **136**, 621 (1982) [Engl. Tranls.: Sov. Phys. Uspekhi **25**, 231 (1982)].
[2] G. Khaliullin and S. Maekawa, Phys. Rev. Lett. **85**, 3950 (2000).
[3] Y. Tokura and N. Nagaosa, Science **288**, 462 (2000).
[4] E. Saitoh, S. Okamoto, K. T. Takahashi, K. Tobe, K. Yamamoto, T. Kimura, S. Ishihara, S. Maekawa, and Y. Tokura, Nature **410**, 180 (2001).
[5] K. Kikoin, O. Entin-Wohlman, V. Fleurov, and A. Aharony, Phys. Rev. B **67**, 214418 (2003).
[6] D. V. Efremov, J. van den Brink, and D. I. Khomskii, Nature Mater. **3**, 853 (2004).
[7] M. B. Salamon and M. Jaime, Rev. Mod. Phys. **73**, 583 (2002).
[8] E. L. Nagaev, *Colossal Magnetoresistance and Phase Separation in Magnetic Semiconductors* (Imperial College Press, London, 2002).
[9] V. Perebeinos and P. B. Allen, Phys. Stat. Sol. (b) **215**, 607 (1999).
[10] D. J. Huang, C. F. Chang, H.-T. Jeng, G. Y. Guo, H.-J. Lin, W. B. Wu, H. C. Ku, A. Fujimori, Y. Takahashi, and C. T. Chen, Phys. Rev. Lett. **93**, 077204 (2004).
[11] J. van den Brink, W. Stekelenburg, D. I. Khomskii, G. A. Sawatzky, and K.I.Kugel, Phys. Rev. B**58**, 10276 (1998).
[12] *Proceedings of the International Meeting on Magnetite and Other Materials Showing a Verwey Transition* [Philos. Mag. B **42**, No. 3, 1980 (special issue)].
[13] G. Subias, J. Garcia, J. Blasco, M. G. Proietti, H. Renevier, and M. C. Sanchez, Phys. Rev. Lett. **93**, 156408 (2004).
[14] V. Yu. Irkhin and M. I. Katsnelson, Phys. Lett. A **150**, 47 (1990).
[15] V. Yu. Irkhin and M. I. Katsnelson, Usp. Fiz. Nauk **164**, 705 (1994) [Engl. Transl.: Physics Uspekhi **37**, 659 (1994)].
[16] V. Yu. Irkhin, M. I. Katsnelson, and A. I. Lichtenstein, cond-mat/0406487 (to be published in Proceedings of the workshop "Local-Moment Ferromagnets: Unique Properties for Modern Applications", Berlin 2004).
[17] V. Yu. Irkhin and Yu. P. Irkhin, Phys. Stat. Sol.(b) **183**, 9 (1994); cond-mat/9812072.
[18] J. Hubbard, Proc. Roy. Soc. A **285**, 542 (1965).
[19] V. Yu. Irkhin and Yu. P. Irkhin, Zh. Eksp. Teor. Fiz. **107**, 616 (1995) [Engl. Transl.: JETP **80**, 334 (1995)].
[20] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, 1993).
[21] A. M. Tsvelick and P. B. Wiegmann, Adv. Phys. **32**, 745 (1983).
[22] Yu. P. Irkhin, Usp. Fiz. Nauk **154**, 321 (1988) [Engl. Transl.: Sov. Phys. Usp.**31**, 163 (1988)].
[23] V. Yu. Irkhin and M. I. Katsnelson, J. Phys. C **18**, 4173 (1985); V. Yu. Irkhin and M. I. Katsnelson, Zh. Eksp. Teor. Fiz. **88**, 522 (1985) [Engl. Transl.: Sov. Phys. JETP **61**, 306 (1985)].
[24] V. Yu. Irkhin and M. I. Katsnelson, J. Phys.: Cond. Mat. **2**, 7151 (1990).
[25] M. I. Auslender, V. Yu. Irkhin, and M. I. Katsnelson, J. Phys. C **21**, 5521 (1988).
[26] V. Yu. Irkhin and A. V. Zarubin, Phys. Rev. B **70**, 035116 (2004).
[27] D. M. Edwards and J. A. Hertz, J. Phys. F **3**, 2191 (1973).
[28] J. Hubbard, Proc. Roy. Soc. A **276**, 238 (1963).
[29] P. Fulde and M. Loewenhaupt, Adv. Phys. **34**, 589 (1986).
[30] M. I. Auslender and V. Yu. Irkhin, J. Phys. C **18**, 3533 (1985).
[31] R. Blinc and B. Żekś, *Ferroelectrics and Antiferroelectrics. Lattice Dynamics* (North-Holland, Amsterdam, 1984).
[32] M. I. Auslender and V. Yu. Irkhin, Sol. State Commun. **56**, 703 (1985).
[33] V. Yu. Irkhin and M. I. Katsnelson, Eur. Phys. J. B **30**, 481 (2002).
[34] G. Tkachov, E. McCann and V. I. Fal'ko, Phys. Rev. B **65**, 024519 (2002).
[35] R. Wiesendanger, H.-J. Guentherodt, G. Guentherodt, R.J. Gambino, and R. Ruf, Phys. Rev. Lett. **65**, 247 (1990).
[36] M. I. Katsnelson and D. M. Edwards, J. Phys.: Condens. Matter **4**, 3289 (1992).

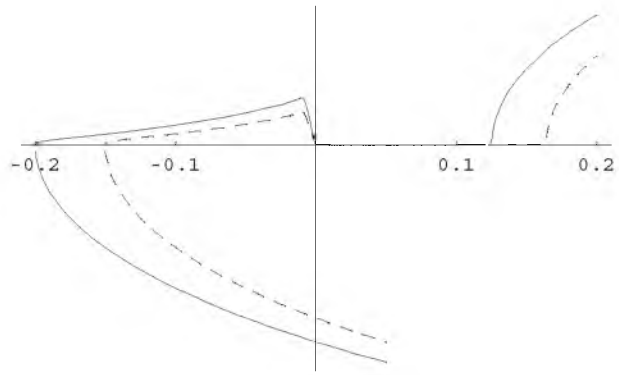

FIG. 1: Density of states in a half-metallic ferromagnet with double orbital degeneracy, $I = -0.3 < 0$ for the semielliptic bare band with the width of $W = 2$, $S = 1/2$, crystal-field splitting $\Delta = 0.1$. The energy is referred to the Fermi level. Solid and dashed lines correspond to crystal-field split subbands, the corresponding Fermi energies calculated from the band bottoms being 0.15 and 0.2, respectively. The spin-down subbands (lower half of the figure) nearly coincide with the bare bands shifted by IS. Non-quasiparticle states in the spin-up subbands (upper half of the figure) occur below the Fermi level.

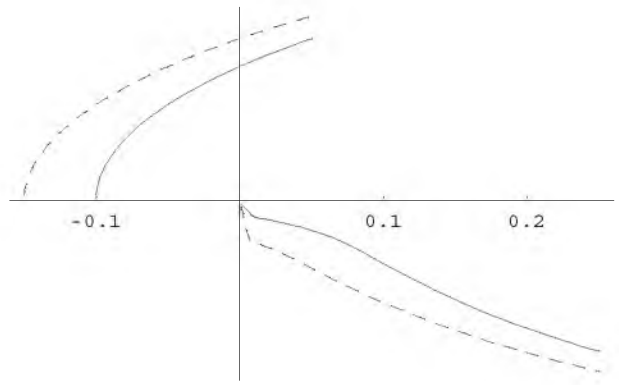

FIG. 2: Density of states in a half-metallic ferromagnet with $I = 0.3 > 0$. The Fermi energies calculated from the orbital subband bottoms are 0.15 and 0.1. Other parameters are the same as in Fig.1 The spin-up subbands (upper half of the figure) coincide with the bare bands shifted by $-IS$. The non-quasiparticle tails of the spin-down subbands (lower half of the figure) occur above the Fermi level.

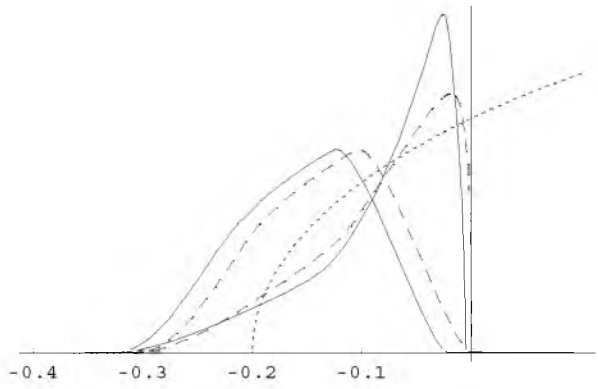

FIG. 3: Density of states $g_{\sigma m}(E)$ in a half-metallic ferromagnet in the model (13). The Fermi energy calculated from the bare band bottom is 0.2 ($c = 0.034$). Short-dashed line is the bare semielliptic density of states ($\sigma = +, m = +$). The solid and dashed lines with peaks near the Fermi level show the contributions from orbital ($\sigma = +$, $m \neq +$, $\Delta = 0.4$) and spin ($\sigma = -, m = +$) channels, respectively. The lower curves (without peaks) show the corresponding functions $-(1/\pi)\mathrm{Im}\overline{\mathrm{R}}_{\sigma\mathrm{m}}(\mathrm{E})$.

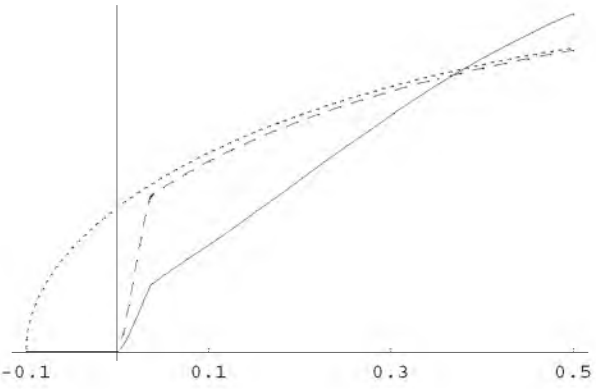

FIG. 4: Density of states in a half-metallic ferromagnet in the model (12). The Fermi energy calculated from the bare band bottom is 0.1 ($c = 0.019$). Solid and short-dashed lines correspond to spin down electrons (the Green's function (53)) and spin up electrons (bare density of states). The dashed line shows the function $-(1/\pi)\mathrm{Im}\overline{\overline{\mathrm{R}}}_{\sigma\mathrm{m}}(\mathrm{E})$.